

EXPERIMENTAL STUDY ON THE MELTING PROCESS OF ICE CRYSTALS JUST BELOW THE MELTING POINT

Takehiko GONDA¹, Tsutomu ARAI² and Tadanori SEI¹

¹ Division of General Education, Aichi Gakuin University, Iwasaki, Nisshin 470-0195

² Seiken Co. Ltd., c/o Nippon Seimei Koishikawa Building, 1-12-14,
Koishikawa, Bunkyo-ku, Tokyo 112-0002

Abstract: The melting process of ice crystals just below the melting point was investigated by *in situ* observation. We found that an ice crystal just below the melting point started to melt at the positions where dislocations or stacking faults emerged on the {0001} and {1010} faces. We also found that the {1010} face of the ice crystal disappeared just below the melting point, while the {0001} face did not disappear even at the melting point.

1. Introduction

There are few studies on the melting of an ice crystal though studies on the surface melting of an ice crystal have been carried out (JACCARD, 1967; FURUKAWA *et al.*, 1989; FURUKAWA and NADA, 1997). GONDA and YAMAZAKI (1980) have studied the dissolution process of an ice crystal in replica solution. In this study, they found that the ice crystal started to melt from corners and then dissolved after its external form resembled a rugby ball like.

SEI and GONDA (1992) observed the melting process of the {0001} and {1010} faces of an ice crystal and the change in shape of these faces with increasing temperature. However, in this study, the details of the melting process and anisotropy of the melting phenomenon could not be determined.

The purpose of this study is to clarify the details of the melting process and the anisotropy of the melting phenomenon by observing simultaneously the {0001} and {1010} faces of the ice crystals.

2. Experiment

Figure 1 shows the growth chamber used in this experiment. The growth chamber was cooled by circulating isopropyl alcohol cooled in advance in a refrigerator (k). Next, the temperature of upper (a) and lower (b) plates was regulated precisely by adjusting an electric current to upper (i) and lower (j) thermoelectric modules. Water vapor was supplied by keeping an ice plate (g) at slightly higher temperature than that of a growth substrate (e). Ice nuclei were supplied by inserting sufficiently diluted silver iodide smoke into the chamber. Ice crystals nucleated in air and fallen on the growth substrate were recorded in a video tape recorder through a video camera using

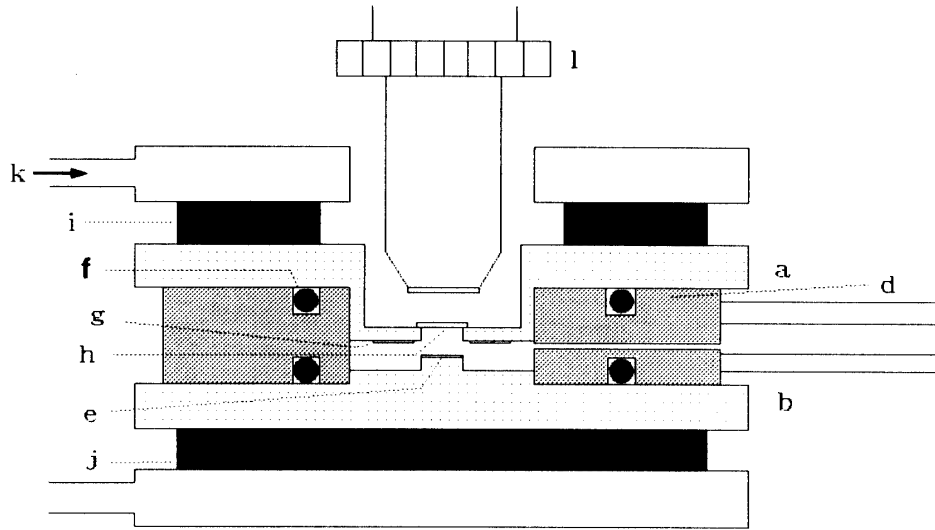


Fig. 1. Growth chamber for ice crystals. a: upper plate, b: lower plate, d: insulator, e: glass substrate, f: o-ring, g: ice plate, h: glass window, i, j: thermoelectric modules, k: coolant, l: objective lens.

a differential interference microscope (l). Here, the substrate temperature and the temperature difference between the substrate and ice plate were measured by connecting C-C thermocouples 0.1 mm in diameter to digital thermometers which can measure six times per second. The accuracy of measured temperatures is $\pm 0.03^\circ\text{C}$. Averages of ten measured values are shown in Figs. 2–4.

3. Experimental Results

In order to study the melting process and anisotropy of ice crystal melting, two crystallographic planes were chosen in the field of view. Ice crystals were formed in air at 6.7×10^3 Pa and -10°C , and then the substrate was heated very slowly to the melting point. As the ice crystals are always in a non-equilibrium state, it is considered that a small amount of heat flows into them over time. Accordingly, as described below, the behavior of the micro drops on ice crystal surfaces changes even when the temperature does not change.

Figure 2 shows the melting process of ice crystals in air at 6.7×10^3 Pa and 0.2% supersaturation when the substrate temperature was heated at the rate of $0.27^\circ\text{C}/\text{min}$. In stage a, both the (0001) and (1010) faces of the ice crystals did not change, but in stage b micro drops appeared along the edges on the (0001) face, where dislocations emerged (GONDA and SEI, 1988).

On the other hand, micro drops also appeared at the positions where stacking faults emerged and at the corner of the (1010) face, where dislocations emerged (arrow \uparrow). In stage c, the size of micro drops on the (0001) face increased, while new micro drops appeared on the (1010) face. In stage d, the micro drops on the (0001) face coagulated with each other and moved irregularly on the (0001) face, while the size of micro drops on (1010) face also increased. In stages e and f, the droplets randomly moved on the

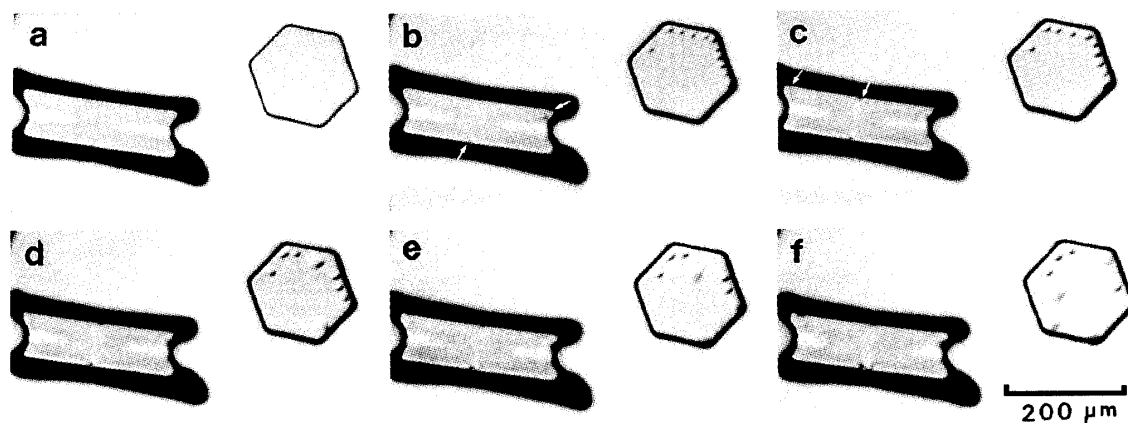


Fig. 2. Melting process of ice crystals in air at 6.7×10^3 Pa and 0.2% supersaturation.
a) -0.47 , b) -0.12 , c) -0.08 , d) -0.07 , e) -0.07 , f) -0.06°C .

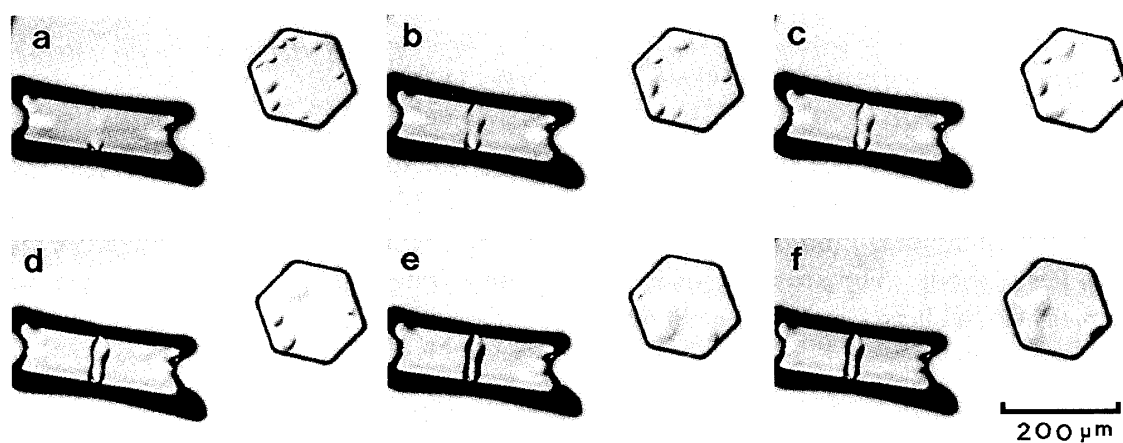


Fig. 3. Further melting of the ice crystals shown in Fig. 2.
a) -0.06 , b) -0.05 , c) -0.05 , d) -0.04 , e) -0.04 , f) -0.03°C .

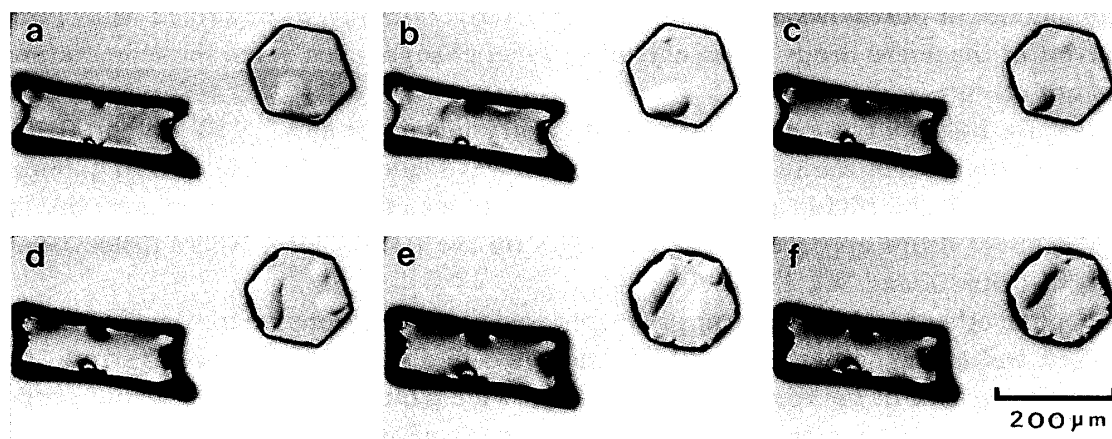


Fig. 4. Further melting of the ice crystals shown in Fig. 3.
a) -0.03 , b) -0.02 , c) -0.02 , d) -0.01 , e) 0.00 , f) 0.00°C .

(0001) face, and the size of droplets on the $\{10\bar{1}0\}$ also increased.

Figure 3 shows further melting of the ice crystals shown in Fig. 2. In stage a, the water droplets on the (0001) and $\{10\bar{1}0\}$ faces coagulated with each other and grew in size.

In stages b, c and d, the same phenomena as stage a occurred. In stages e and f, the droplets randomly moved on the (0001) and $\{10\bar{1}0\}$ faces, respectively and, as a result, the their shapes changed.

Figure 4 shows further melting of the ice crystals shown in Fig. 3. In stage a, the area covered with film-like water drops on the $\{10\bar{1}0\}$ face is larger than that on the (0001) face. This tendency is marked in stage b. In stage c, the $\{10\bar{1}0\}$ face was wholly covered with water film, while the (0001) face was not covered. In stage d, the size of water droplets on the (0001) face increased. Even in stages e and f (melting point), the (0001) face did not disappear. From many experiments, we found that the (0001) and $\{10\bar{1}0\}$ faces of the ice crystals just below the melting point started to melt at the positions where dislocations or stacking faults emerged. Moreover, we also found that the $\{10\bar{1}0\}$ face disappeared just below the melting point, while the (0001) face did not melt even at the melting point.

4. Discussion

Ice crystals started to melt at positions where dislocations or stacking faults emerged on the $\{0001\}$ and $\{10\bar{1}0\}$ faces. The reason is because the chemical potential becomes larger than on a perfect crystal surface by the supplement of strain energy at positions where dislocations or stacking faults emerge.

Figure 5 shows the estimated structure of the $\{0001\}$ and $\{10\bar{1}0\}$ faces of an ice crystal just below the melting point. The fact that the $\{10\bar{1}0\}$ face disappears just below the melting point may be related to roughening at the water/ice interface on the $\{10\bar{1}0\}$ face just below the melting point. On the other hand, the fact that the $\{0001\}$ face does not disappear even at the melting point may be related to a smooth interface with QLL (quasi liquid layer) just below the melting point. However, the precise explanation is uncertain in the present stage.

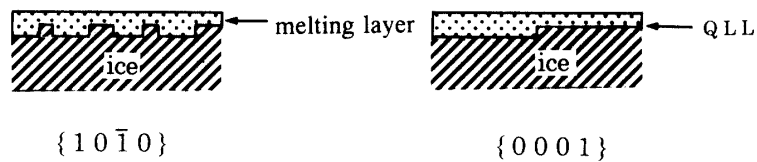


Fig. 5. Estimated structure of the $\{0001\}$ and $\{10\bar{1}0\}$ faces of an ice crystal just below the melting point.

5. Conclusions

The melting process of ice crystals just below the melting point was observed *in situ*. We found that ice crystals started to melt at positions where dislocations or stacking faults emerged on the $\{0001\}$ and $\{10\bar{1}0\}$ faces. The $\{10\bar{1}0\}$ face of ice crystals

disappeared just below the melting point, while the {0001} face did not disappear even at the melting point.

References

- FURUKAWA, Y. and NADA, H. (1997): Anisotropy in microscopic structures of ice-water and ice-vapor interfaces and its relation to growth kinetics. *Advances in the Understanding of Crystal Growth Mechanism*, ed. by T. NISHINAGA *et al.* Elsevier Science B.V., 559–573.
- FURUKAWA, Y., YAMAMOTO, M. and KURODA, T. (1989): Ellipsometric study of the transition layer on the surface of an ice crystal. *J. Crystal Growth*, **82**, 665–67.
- GONDA, T. and SEI, T. (1988): Kisô kara seichô suru kôri no kessho no seichô kikô to katachi (Growth mechanism and the habit change of ice crystals growing from the vapour phase). *Nippon Kessho Seichô Gakkai Shi (J. Jpn. Associ. Crystal Growth)*, **15**, 311–321.
- GONDA, T. and YAMAZAKI, T. (1980): The dissolution and the change in shapes of ice crystals in replica solution. *J. Meteorol. Soc. Jpn.*, **58**, 137–142.
- JACCARD, C. (1967): Electrical conductivity of the surface layer of ice. *Proceedings of the International Conference Low Temperature Science*, Vol. 1, ed. by H. ÔURA. Sapporo, Inst. Low Temp. Sci., Hokkaido Univ., 173–179.
- SEI, T. and GONDA, T. (1992): Faceted growth of ice crystals growing at temperatures just below the melting point. *Physics and Chemistry of Ice*, ed. by N. MAENO and T. HONDO. Sapporo, Hokkaido Univ. Press, 349–356.

(Received April 8, 1998; Revised manuscript accepted September 14, 1999)